FORM PTO-1390 (Modified) (REV 11-2000) U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NUMBER	
YPO0031	
U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR)	09/980225

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/JP00/05131	07/31/2000	07/30/1999

TITLE OF INVENTION

MOLDING MATERIAL FOR USE WITH CARBON DIOXIDE REFRIGERANT

APPLICANT(S) FOR DO/EO/US

OSAMU KOBAYASHI ET AL.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include itens (5), (6), (9) and (24) indicated below.
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371 (c) (2))
   a. ☐ is attached hereto (required only if not communicated by the International Bureau).
   b. ☒ has been communicated by the International Bureau.
   c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
   a. ☒ is attached hereto.
   b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))
   a. ☐ are attached hereto (required only if not communicated by the International Bureau).
   b. ☐ have been communicated by the International Bureau.
   c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
   d. ☐ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).
10. ☒ An English language translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).
11. ☒ A copy of the International Preliminary Examination Report (PCT/IPEA/409).
12. ☒ A copy of the International Search Report (PCT/ISA/210).

Items 13 to 20 below concern document(s) or information included:

13. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
14. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
15. ☒ A **FIRST** preliminary amendment.
16. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
17. ☐ A substitute specification.
18. ☐ A change of power of attorney and/or address letter.
19. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
20. ☒ A second copy of the published international application under 35 U.S.C. 154(d)(4).
21. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
22. ☒ Certificate of Mailing by Express Mail
23. ☐ Other items or information:

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR)	INTERNATIONAL APPLICATION NO.	ATTORNEY'S DOCKET NUMBER
09/980225	PCT/JP00/05131	YPO0031

24. The following fees are submitted:.	CALCULATIONS PTO USE ONLY
BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) :	
☐ Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO **$1040.00**	
☒ International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO **$890.00**	
☐ International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO **$740.00**	
☐ International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) **$710.00**	
☐ International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) **$100.00**	
ENTER APPROPRIATE BASIC FEE AMOUNT =	**$890.00**
Surcharge of **$130.00** for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492 (e)).	**$0.00**

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	
Total claims	12 - 20 =	0	x $18.00	**$0.00**
Independent claims	1 - 3 =	0	x $84.00	**$0.00**
Multiple Dependent Claims **(check if applicable)**.			☐	**$0.00**
TOTAL OF ABOVE CALCULATIONS =				**$890.00**
☐ Applicant claims small entity status. See 37 CFR 1.27). The fees indicated above are reduced by 1/2.				**$0.00**
SUBTOTAL =				**$890.00**
Processing fee of **$130.00** for furnishing the English translation later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492 (f)). +				**$0.00**
TOTAL NATIONAL FEE =				**$890.00**
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) **(check if applicable).** ☒				**$40.00**
TOTAL FEES ENCLOSED =				**$930.00**
				Amount to be: refunded $
				charged $

a. ☒ A check in the amount of **$930.00** to cover the above fees is enclosed.

b. ☐ Please charge my Deposit Account No. _____ in the amount of _____ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. **02-0385** A duplicate copy of this sheet is enclosed.

d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. **Credit card information should not be included on this form.** Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

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Michael S. Gzybowski
SIGNATURE

Michael S. Gzybowski
NAME

32,816
REGISTRATION NUMBER

November 29, 2001
DATE

PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Group Art Unit:	Unknown
Attorney Docket No.:	YPO0031
Applicant:	Osamu Kobayashi et al.
Invention:	MOLDING MATERIAL FOR USE WITH CARBON DIOXIDE REFRIGERANT
U.S. Serial No:	Unknown
U.S. Filing Date:	Herewith
Int'l Serial No.:	PCT/JP00/05131
Int'l Filing Date:	July 31, 2000
Examiner:	Unknown

Certificate Under 37 CFR 1 10

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DATE OF DEPOSIT NOVEMBER 29, 2001
I HEREBY CERTIFY THAT THIS PAPER OR FEE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE "EXPRESS MAIL POST OFFICE TO ADDRESSEE" SERVICE UNDER 37 CFR 1.10 ON THE DATE INDICATED ABOVE AND IS ADDRESSED TO THE ASSISTANT COMMISSIONER FOR PATENTS WASHINGTON, DC 20231

on NOVEMBER 29, 2001

Michelle L. Neal

PRELIMINARY AMENDMENT

Box New Application
Attn: DO/EO/US
Assistant Commissioner for Patents
Washington, DC 20231

Sir:

Prior to the examination of the above-identified application, please amend the application as follows:

IN THE TITLE

Please change the title from "MOLDING MATERIAL FOR CARBON DIOXIDE" to - -MOLDING MATERIAL FOR USE WITH CARBON DIOXIDE REFRIGERANT- -

IN THE SPECIFICATION

Please replace the first full paragraph on page 1 with the following:

- -The present invention relates to a molding material for use with carbon dioxide refrigerant, and more particularly to a molding material for use with carbon dioxide refrigerant with distinguished resistance to carbon dioxide gas permeation and volumic swelling.- -

Please replace the second full paragraph on page 2 with the following:

- -An object of the present invention is to provide a molding material for use with carbon dioxide refrigerant, which can be used as suitable molding materials for sealing materials, etc. in carbon dioxide refrigerant-contacting apparatuses.- -

Please replace the third full paragraph on page 2 with the following:

- -The object of the present invention can be attained by a molding material for use with carbon dioxide refrigerant, which comprises chlorinated polyethylene having a chlorine content of 25-47% by weight. A blend of chlorinated polyethylene with vinyl chloride-based resin can be also used. furthermore, it is particularly effective for prevention of blister generation to add a specific silane coupling agent thereto.- -

Please replace the first full paragraph on page 11 with the following:

- -The present molding materials for use with carbon dioxide refrigerant with the foregoing properties can be used as suitable materials for sealing materials such as packings, gaskets, O-rings, etc. in carbon dioxide refrigerant-contacting apparatuses such as air compressors, refrigerators, supercritical CO_2 extraction(chromatography) apparatuses, etc. using carbon dioxide as a refrigerant, and also as suitable materials for sealing materials for refrigerator oil (polyalkylene gycol, etc.) applicable to the carbon dioxide refrigerant.- -

IN THE CLAIMS

Please amend claims 1-6 as follows:

1. (Amended) A molding material for use with carbon dioxide refrigerant, which comprises chlorinated polyethylene having a chlorine content of 25-47% by weight.

2. (Amended) A molding material for use with carbon dioxide refrigerant according to Claim 1, wherein the chlorinated polyethylene is used as a blend with vinyl chloride-based resin.

3. (Amended) A molding material for use with carbon dioxide refrigerant according to Claim 1, wherein an inorganic filler is further contained.

4. (Amended) A molding material for use with carbon dioxide refrigerant according to Claim 1, wherein a vinylic, epoxy or methacryloxy silane coupling agent is further contained.

5. (Amended) A molding material for use with carbon dioxide refrigerant according to Claim 1, wherein 1-10 parts by weight of an organic peroxide is further contained on the basis of 100 parts by weight of the chlorinated polyethylene or its blend with the vinyl chloride-based resin.

6. (Amended) A molding material for use with carbon dioxide refrigerant according to Claim 1, for use as a molding material for sealing materials in carbon dioxide refrigerant-contacting apparatuses.

Please add new claim 7 as follows:

- -7. A molding material for use with carbon dioxide refrigerant according to Claim 2, wherein a vinylic, epoxy or methacryloxy silane coupling agent is further contained.- -

Please add new claim 8 as follows:

- -8. A molding material for use with carbon dioxide refrigerant according to Claim 2, wherein an inorganic filler is further contained.- -

Please add new claim 9 as follows:

- -9. A molding material for use with carbon dioxide refrigerant according to Claim 2, wherein 1-10 parts by weight of an organic peroxide is further contained on the basis of 100 parts by weight of the chlorinated polyethylene or its blend with the vinyl chloride-based resin.- -

Please add new claim 10 as follows:

- -10. A molding material for use with carbon dioxide refrigerant according to Claim 4, wherein 1-10 parts by weight of an organic peroxide is further contained on the basis of 100 parts by weight of the chlorinated polyethylene or its blend with the vinyl chloride-based resin.- -

Please add new claim 11 as follows:

- -11. A molding material for use with carbon dioxide refrigerant according to Claim 2, for use as a molding material for sealing materials in carbon dioxide refrigerant-contacting apparatuses.- -

Please add new claim 12 as follows:

- -12. A molding material for use with carbon dioxide refrigerant according to Claim 4, for use as a molding material for sealing materials in carbon dioxide refrigerant-contacting apparatuses.- -

• • • REMARKS • • •

By the present Preliminary Amendment, the title has been amended to reflect the title in the English translation of the specification.

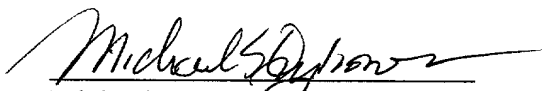
The specification and claims have been amended to include the changes made to the international application as amended under Article 34.

The claims have also been amended to delete multiple dependency.

Entry of the present Preliminary Amendment prior to the examination of the application is respectfully requested.

In the event Applicants have overlooked the need for an extension of time, an additional extension of time, payment of fee, or additional payment of fee, Applicants hereby petition therefor and authorize that any charges be made to Deposit Account No. 02-0385, Baker & Daniels.

Respectfully submitted,



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VERSION WITH MARKINGS TO SHOW CHANGES MADE

Changes Made to Specification Paragraphs

The first full paragraph on page 1 has been amended as follows:

The present invention relates to a molding material for use with carbon dioxide refrigerant, and more particularly to a molding material for use with carbon dioxide refrigerant with distinguished resistance to carbon dioxide gas permeation and volumic swelling.

The second full paragraph on page 2 has been amended as follows:

An object of the present invention is to provide a molding material for use with carbon dioxide refrigerant, which can be used as suitable molding materials for sealing materials, etc. in carbon dioxide refrigerant-contacting apparatuses.

The third full paragraph on page 2 has been amended as follows:

The object of the present invention can be attained by a molding material for use with carbon dioxide refrigerant, which comprises chlorinated polyethylene having a chlorine content of 25-47% by weight. A blend of chlorinated polyethylene with vinyl chloride-based resin can be also used. furthermore, it is particularly effective for prevention of blister generation to add a specific silane coupling agent thereto.

The first full paragraph on page 11 has been amended as follows:

The present molding materials for use with carbon dioxide refrigerant with the foregoing properties can be used as suitable materials for sealing materials such as packings, gaskets, O-rings, etc. in carbon dioxide refrigerant-contacting apparatuses such as air compressors, refrigerators, supercritical CO_2 extraction(chromatography) apparatuses, etc. using carbon dioxide as a refrigerant, and also as suitable materials for sealing materials for refrigerator oil (polyalkylene gycol, etc.) applicable to the carbon dioxide refrigerant.

The first full paragraph on page 1 has been amended as follows:

The present invention relates to a molding material for use with carbon dioxide refrigerant, and more particularly to a molding material for use with carbon dioxide refrigerant with distinguished resistance to carbon dioxide gas permeation and volumic swelling.

The second full paragraph on page 2 has been amended as follows:

An object of the present invention is to provide a molding material for use with carbon dioxide refrigerant, which can be used as suitable molding materials for sealing materials, etc. in carbon dioxide refrigerant-contacting apparatuses.

The third full paragraph on page 2 has been amended as follows:

The object of the present invention can be attained by a molding material for use with carbon dioxide refrigerant, which comprises chlorinated polyethylene having a chlorine content of 25-47% by weight. A blend of chlorinated polyethylene with vinyl chloride-based resin can be also used. furthermore, it is particularly effective for prevention of blister generation to add a specific silane coupling agent thereto.

The first full paragraph on page 11 has been amended as follows:

The present molding materials for use with carbon dioxide refrigerant with the foregoing properties can be used as suitable materials for sealing materials such as packings, gaskets, O-rings, etc. in carbon dioxide refrigerant-contacting apparatuses such as air compressors, refrigerators, supercritical CO_2 extraction(chromatography) apparatuses, etc. using carbon dioxide as a refrigerant, and also as suitable materials for sealing materials for refrigerator oil (polyalkylene gycol, etc.) applicable to the carbon dioxide refrigerant.

Changes Made to Claims

Claim 1 has been amended as follows:

1. (Amended) A molding material for use with carbon dioxide refrigerant, which comprises chlorinated polyethylene having a chlorine content of 25-47% by weight.

Claim 2 has been amended as follows:

2. (Amended) A molding material for use with carbon dioxide refrigerant according to Claim 1, wherein the chlorinated polyethylene is used as a blend with vinyl chloride-based resin.

Claim 3 has been amended as follows:

3. (Amended) A molding material for use with carbon dioxide refrigerant according to Claim 1 [or 2], wherein an inorganic filler is further contained.

Claim 4 has been amended as follows:

4. (Amended) A molding material for use with carbon dioxide refrigerant according to Claim 1 [or 2], wherein a vinylic, epoxy or methacryloxy silane coupling agent is further contained.

Claim 5 has been amended as follows:

5. (Amended) A molding material for use with carbon dioxide refrigerant according to Claim 1, [2 or 4,] wherein 1-10 parts by weight of an organic peroxide is further contained on the basis of 100 parts by weight of the chlorinated polyethylene or its blend with the vinyl chloride-based resin.

Claim 6 has been amended as follows:

6. (Amended) A molding material for use with carbon dioxide refrigerant according to Claim 1, [2 or 4] for use as a molding material for sealing materials in carbon dioxide refrigerant-contacting apparatuses.

Claim 7 has been added as follows:

7. A molding material for use with carbon dioxide refrigerant according to Claim 2, wherein a vinylic, epoxy or methacryloxy silane coupling agent is further contained.

Claim 8 has been added as follows:

8. A molding material for use with carbon dioxide refrigerant according to Claim 2, wherein an inorganic filler is further contained.

Claim 9 has been added as follows:

9. A molding material for use with carbon dioxide refrigerant according to Claim 2, wherein 1-10 parts by weight of an organic peroxide is further contained on the basis of 100 parts by weight of the chlorinated polyethylene or its blend with the vinyl chloride-based resin.

Claim 10 has been added as follows:

10. A molding material for use with carbon dioxide refrigerant according to Claim 4, wherein 1-10 parts by weight of an organic peroxide is further contained on the basis of 100 parts by weight of the chlorinated polyethylene or its blend with the vinyl chloride-based resin.

Claim 11 has been added as follows:

11. A molding material for use with carbon dioxide refrigerant according to Claim 2, for use as a molding material for sealing materials in carbon dioxide refrigerant-contacting apparatuses.

Claim 12 has been added as follows:

12. A molding material for use with carbon dioxide refrigerant according to Claim 4, for use as a molding material for sealing materials in carbon dioxide refrigerant-contacting apparatuses.

AMENDED SPECIFICATION (UNDER ARTICLE 34)

MOLDING MATERIAL FOR USE WITH CARBON DIOXIDE REFRIGERANT

TECHNICAL FIELD

The present invention relates to a molding material for use with carbon dioxide refrigerant, and more particularly to a molding material for use with carbon dioxide refrigerant with distinguished resistance to carbon dioxide gas permeation and volumic swelling.

BACKGROUND ART

Freon R-134a (1,1,1,2-tetrafluoroethane) called a new refrigerant is now mainly used in refrigerators, etc., but it is expected that its use would be prohibited in future in view of problems of environmental control, etc. Hydrocarbon gases and carbon dioxide are now regarded as important substitutes for such a refrigerant. Hydrocarbon gases have a considerably high risk of explosion or combustion and the world trend toward the next generation refrigerant is shifting to carbon dioxide.

Carbon dioxide requires a higher pressure than freon and also has higher permeability and solubility into ordinary polymer materials and thus has a higher chance of blister (foaming) generation. Even if there is no blister generation, carbon dioxide gas permeates through the polymer materials, resulting in failure of pressure maintenance or tight sealing.

Even rubber materials have a high carbon dioxide gas permeability in general, and the permeability is considerably high particularly under pressure of 10 atm or higher, resulting in failure of tight sealing of carbon dioxide gas. Furthermore, carbon dioxide is very soluble into polymers,

giving rise to considerable polymer swelling. Thus, no rubber material parts have been actually applied to apparatuses using carbon dioxide.

In case of the conventional freon gas refrigerants, hydrogenated NBR, EPDM, etc. have been used as molding materials for sealing materials, but these rubber materials considerably swell or blister through contact with carbon dioxide as disadvantages.

DISCLOSURE OF THE INVENTION

An object of the present invention is to provide a molding material for use with carbon dioxide refrigerant, which can be used as suitable molding materials for sealing materials, etc. in carbon dioxide refrigerant-contacting apparatuses.

The object of the present invention can be attained by a molding material for use with carbon dioxide refrigerant, which comprises chlorinated polyethylene having a chlorine content of 25-47% by weight. A blend of chlorinated polyethylene with vinyl chloride-based resin can be also used. Furthermore, it is particularly effective for prevention of blister generation to add a specific silane coupling agent thereto.

Chlorinated polyethylene for use in the present invention has a chlorine content of 25-47% by weight, preferably 28-45% by weight. The lower the chlorine content than 25% by weight, the nearer to the property of polyethylene, resulting in loss of rubber elasticity. Sealing materials molded from such chlorinated polyethylene will have poor sealing. With a higher chlorine content than 47% by weight, the low temperature resistance, heat resistance, etc. will be lowered. Actually, commercial products with the above-mentioned range of chlorine content, for example, Elaslene series of Showa Denko products, can be used as such.

Chlorinated polyethylene with such a range of chlorine content can be usually used alone, but in case of improving vulcanization product properties such as tensile strength, elongation, etc. at somewhat the sacrifice of resistances to carbon dioxide gas permeation and volume swelling, a blend of chlorinated polyethylene with vinyl chloride-based resin, usually polyvinyl chloride, can be also used, where a blending proportion of vinyl chloride-based resin is not more than about 50% by weight, preferably about 10 to about 40% by weight, on the basis of the blend.

From the viewpoint of dynamic properties and resistance to gas permeation of chlorinated polyethylene or its blend, about 5 to about 150 parts by weight, preferably about 10 to about 100 parts by weight, of an inorganic filler is added to 100 parts by weight of chlorinated polyethylene or its blend. Reinforcing fillers such as carbon black, silica, etc. can be usually used as an inorganic filler. Furthermore, non-reinforcing fillers such as calcium metasilicate, diatomoceous earth, graphite, mica, calcium carbonate, zinc oxide, etc. can be also used together with the reinforcing fillers.

Furthermore, an organic peroxide is added as a cross-linking agent to the molding material comprising the above-mentioned components. Organic peroxide for use in the present invention includes, for example, 1,1-bis(t-butylperoxy)-3,5,5-trimethylcyclo-hexane, 2,5-dimethylhexane-2,5-dihydroxyperoxide, di-t-butyl peroxide, t-butyl cumyl peroxide, dicumyl peroxide, t-butylperoxyisopropyl carbonate, n-butyl-4,4-di(t-butylperoxy) valerate, α , α -bis(t-butylperoxy)-p-diisopropylbenzene, 1,3-di(t-butyl-peroxyisopropyl)benzene, 2,5-dimethyl-2,5-di(t-butylperoxy)hexane, 2,5-dimethyl-2,5-di(t-butylperoxy)hexine-3, benzoyl peroxide, t-butylperoxy-

benzene, 2,5-dimethyl-2,5-di(benzoylperoxy)hexane, etc., and is used in a proportion of about 1 to about 10 parts by weight, preferably about 2 to about 8 parts by weight on the basis of 100 parts by weight of chlorinated polyethylene or its blend. In a lower proportion of organic peroxide than about 1 part by weight no satisfactory cross-linking density can be obtained, whereas in a higher proportion than about 10 parts by weight no vulcanization moldings can be obtained due to foaming, or rubber elasticity, elongation or compression set characteristics will be inevitably lowered, even if moldings can be obtained.

To the molding material containing an organic peroxide as a cross-linking agent it is added about 0.1 to about 10 parts by weight, preferably about 1 to about 5 parts by weight, of a polyfunctional unsaturated compound, typically triallyl isocyanurate, triallyl cyanurate, etc. as a cocross-linking agent on the basis of 100 parts by weight of chlorinated polyethylene or its blend. When the cross-linking agent and the cocross-linking agent are used in higher proportions than about 10 parts by weight, respectively, scorching, etc. will take place.

Furthermore, it is effective for prevention of blister generation due to contact with carbon dioxide to add at least 0.2 parts by weight, preferably about 0.5 to about 5 parts by weight, of a specific silane coupling agent to 100 parts by weight of chlorinated polyethylene or its blend with vinyl chloride-based resin.

Such a specific silane coupling agent includes, for example, vinylic silane coupling agents such as vinyltrimethoxysilane, vinyltriethoxysilane, vinyltris(2-methoxyethoxy)silane, vinyltrichlorosilane, etc., epoxy silane coupling agents such as γ -glycidoxypropyltrimethoxysilane, γ -glycidoxypropyltriethoxysilane, γ -glycidoxypropylmethyldiethoxysilane,

etc., and methacryloxysilane coupling agents such as γ -methacryloxypropyltrimethoxysilane, γ -methacryloxypropyltriethoxysilane, γ -methacryloxypropylmethyldimethoxysilane, γ -methacryloxypropylmethyldiethoxysilane, etc.

Chlorinated polyethylene composition comprising the foregoing components can be prepared by further adding thereto, if required, various additives, for example, a processing aid such as stearic acid, palmitic acid, paraffin wax, etc., an acid acceptor such as zinc oxide, magnesium oxide, etc., an antioxidant, a plasticizer, etc. properly, followed by kneading through Intermix, kneader, Banbury mixer, etc. or through open rolls.

Vulcanization of the composition is carried out usually by heating at about 150° to about 200°C for about 3 to about 60 minutes though a vulcanization press, an injection molding machine, a compression molding machine, etc., and, if required, secondary vulcanization is carried out by heating at about 120° to about 200°C for about 1 to about 24 hours.

BEST MODE FOR CARRYING OUT THE INVENTION

The present invention will be described below, referring to Examples.

EXAMPLE 1

	Parts by weight
Chlorinated polyethylene (Elaslene 302NA, product by Showa Denko K. K.; chlorine content: 29 wt.%)	100
Carbon black (Thermax N990, product by Cancarb)	120
Magnesium oxide (Kyowa Mag #150, product by Kyowa Kagaku K. K.)	5
Organic peroxide (Percumyl D. product by NOF	5

Corp.)

Triallyl isocyanurate (Tyke, product by Nihon Kasei K. K.) 3

The foregoing components were kneaded through 10-inch open rolls (roll temperature : 130°C), and the kneading product was press vulcanized at 180°C for 6 minutes to obtain a vulcanization molded circular sheet, 90mm in diameter and 0.5mm thick.

A disc sample, 50mm in diameter, was cut out from the circular sheet and mounted on a high pressure gas permeability tester to determine a CO_2 permeability under high pressure (60 atm). It was found to be $2.7\times 10^{-10}cm^3$ (STP) · cm/cm^2 · sec · cmHg. Furthermore, a rectangular sample, 20mm×5mm, was cut out from the circular sheet and exposed to a CO_2 atmosphere under high pressure (60 atm) in a pressurizing apparatus with a window. A volumic swelling rate $\triangle V$ was measured by a cathetometer and found to be 10 vol.%.

EXAMPLE 2

In Example 1, the same amount of Elaslene 406AE, a product by Showa Denko K. K. (chlorine content : 39.5 wt.%) as chlorinated polyethylene and 70 parts by weight of Seest G-S, product by Tokai Carbon Co., Ltd. as carbon black were used. It was found that CO_2 permeability was $4.8\times 10^{-10}cm^3$ (STP) · cm/cm^2 · sec · cmHg and volumic swelling rate $\triangle V$ was 11 vol.%.

EXAMPLE 3

In Example 2, the amount of carbon black was changed to 50 parts by weight and 40 parts by weight of Nipseal ER, product by Nippon Silica K. K. was additionally used as white carbon. It was found that CO_2 permeability was $3.0\times 10^{-10}cm^3$ (STP) · cm/cm^2 · sec · cmHg and volumic

swelling rate $\triangle V$ was 10 vol.%.

EXAMPLE 4

In Example 1, the same amount of Elaslene 452NA, product by Showa Denko K. K. (chlorine content : 44.5 wt.%) was used as chlorinated polyethylene and the amount of carbon black was changed to 90 parts by weight. It was found that CO_2 permeability was 2.2×10^{-10} cm^3 (STP) · cm/cm^2 · sec · cmHg and volumic swelling rate $\triangle V$ was 9 vol.%.

EXAMPLE 5

In Example 1, the amount of chlorinated polyethylene was changed to 70 parts by weight and 30 parts by weight of polyvinyl chloride (ZEST800Z, product by Shin-Daiichi Enbi K. K.) was used as a blend with the chlorinated polyethylene. It was found that CO_2 permeability was 7.6×10^{-10} cm^3 (STP) · cm/cm^2 · sec · cmHg and volumic swelling rate $\triangle V$ was 16 vol.%.

COMPARATIVE EXAMPLE 1

	Parts by weight
Hydrogenated NBR (Zetpol 1020, product by Nippon Zeon Co., Ltd.)	100
Carbon black (Thermax N990)	80
Magnesium oxide (Kyowa Mag #150)	5
Organic peroxide (Percumyl D)	5
Triallyl isocyanurate (Tyke)	3

The foregoing components were subjected to kneading, vulcanization, molding and determination in the same manner as in Example 1, and it was found that CO_2 permeability was 7.3×10^{-9} cm^3 (STP) · cm/cm^2 · sec · cmHg and volumic swelling rate $\triangle V$ was 31 vol.%.

Vulcanization molding products of the foregoing Examples and

Comparative Example were subjected to determination of normal state physical properties according to JIS K-6301. The results are give in the following Table 1.

Table 1

Example No.	Tensile strength (MPa)	Elongation (%)
1	15	200
2	16	210
3	17	190
4	17	205
5	22	275
Comp. Ex. 1	18	250

EXAMPLE 6

	Parts by weight
Chlorinated polyethylene (Elaslene 352NA, product by showa Denko K. K., chlorine content : 35 wt.%)	100
SRF carbon black	55
Magnesium oxide	5
Dicumyl peroxide	4
Triallyl isocyanurate	5
Vinylic silane coupling agent (vinyl triethoxysilane)	2

The foregoing components were kneaded through a kneader and open rolls, and the kneading product was press vulcanized at 170℃ for 30 minutes and then oven vulcanized (second vulcanization) at 140℃ for 10 hours to obtain a vulcanization sheet, 150mm×150mm×2mm.

EXAMPLE 7

In Example 6, the same amount of epoxy silane coupling agent (γ-glycidoxypropyltriethoxysilane) was used in place of the vinylic silane coupling agent.

EXAMPLE 8

In Example 6, the same amount of methacryloxy silane coupling agent (γ-methacryloxypropyltriethyoxysilane) was used in place of the vinylic silane coupling agent.

COMPARATIVE EXAMPLE 2

In Example 6, the amount of dicumyl peroxide was changed to 0.5 parts by weight.

COMPARATIVE EXAMPLE 3

In Example 6, the amount of dicumyl peroxide was changed to 12 parts by weight.

COMPARATIVE EXAMPLE 4

In Example 6, the same amount of amino silane coupling agent (γ-aminopropyltriethoxysilane) was used in place of the vinylic silane coupling agent.

Vulcanization sheets obtained in the foregoing Examples 6 to 8 and Comparative Examples 2 to 4 were subjected to determination and evaluation of the following items. Results are shown in Table 2. In the case of Comparative Example 3, no vulcanization molding could be carried out due to foaming

Normal state physical properties: according to JIS K-6253 and JIS K-6251

Compression set: according to JIS K-6262

CO_2 resistance: The sheets were dipped into liquidified CO_2 at

25℃ for 24 hours, followed by heating at 150℃ for one hour and then by visual inspection of blister generation on the sheet surface

Table 2

Items of determination and evaluation	Ex.6	Ex.7	Ex.8	Comp. Ex.2	Comp. Ex.4
[Normal state physical properties]					
Hardness (durometer A)	85	85	84	74	85
Tensile strength (MPa)	20.0	20.2	20.7	24.8	17.9
Elongation (%)	200	210	190	500	350
[Compression set]					
120℃ for 70 hours (%)	20	20	18	67	43
[CO_2 resistance]					
Blister generation	None	None	None	None	Occurred

INDUSTRIAL APPLICATION

Molding materials for use with carbon dioxide refrigerant, which comprise chlorinated polyethylene, have a CO_2 permeability under 60 atm in the order of $10^{-10}cm^3$ (STP) · cm/cm^2 · sec · cmHg or less and a distinguished volumic swelling rate $\triangle V$ of about 10 vol. % or less. When chlorinated polyethylene is used as a blend with vinyl chloride-bassed resin, good vulcanization product properties such as tensile strength : 18 MPa or higher and elongation : 250% or higher can be obtained, though resistances to carbon dioxide gas permeation and volumic swelling may be somewhat sacrificed. Furthermore, when a specific silane coupling agent is added to

chlorinated polyethylene or its blend with vinyl chloride-based resin, blister generation can be effectively prevented.

The present molding materials for use with carbon dioxide refrigerant with the foregoing properties can be used as suitable materials for sealing materials such as packings, gaskets, O-rings, etc. in carbon dioxide refrigerant -contacting apparatuses such as air compressors, refrigerators, supercritical CO_2 extraction (chromatography) apparatuses, etc. using carbon dioxide as a refrigerant, and also as suitable materials for sealing materials for refrigerator oil (polyalkylene glycol, etc.) applicable to the carbon dioxide refrigerant.

CLAIMS

1. (AMENDED) A molding material for use with carbon dioxide refrigerant, which comprises chlorinated polyethylene having a chlorine content of 25-47% by weight.

2. (AMENDED) A molding material for use with carbon dioxide refrigerant according to Claim 1, wherein the chlorinated polyethylene is used as a blend with vinyl chloride-based resin.

3. (AMENDED) A molding material for use with carbon dioxide refrigerant according to Claim 1 or 2, wherein an inorganic filler is further contained.

4. (AMENDED) A molding material for use with carbon dioxide refrigerant according to Claim 1 or 2, wherein a vinylic, epoxy or methacryloxy silane coupling agent is further contained.

5. (AMENDED) A molding material for use with carbon dioxide refrigerant according to Claim 1, 2 or 4, wherein 1-10 parts by weight of an organic peroxide is further contained on the basis of 100 parts by weight of the chlorinated polyethylene or its blend with the vinyl chloride-based resin.

6. (AMENDED) A molding material for use with carbon dioxide refrigerant according to Claim 1, 2 or 4 for use as a molding material for sealing materials in carbon dioxide refrigerant-contacting apparatuses.

ATTORNEY'S DOCKET NO. ~~SHC01~~ YP00081

PCT/USA NATIONAL DECLARATION AND POWER OF ATTORNEY FOR U.S. PATENT APPLICATIONS IN THE UNITED STATES PATENT AND TRADEMARK OFFICE UNDER 35 U.S.C. SECTION 371(c)(4)

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name:

I verily believe I am the original, first and sole inventor (if only one name is listed below) or a joint inventor (if plural inventors are named below) of the invention described and claimed in international application No. PCT/JP00/05131 **entitled:**

MOLDING MATERIAL FOR USE WITH CARBON DIOXIDE

and as amended on January 23, 2001 **(if any), which I have reviewed, and I understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above and for which I solicit a patent; that I do not know and do not believe that this invention was ever known or used in the United States of America before my or our invention or discovery thereof, or patented or described in any printed publication in any country before my or our invention or discovery thereof, or more than one year prior to my international application; that this invention was not in public use or on sale in the United States of America for more than one year prior to my international application; that this invention has not been patented or made the subject of an inventor's certificate issued before the date of my international application in any country foreign to the United States of America on an application filed by me or my legal representatives or assigns more than twelve months before my international application; that I acknowledge my duty to disclose information of which I am aware which is material to patentability of this application; and that prior to filing said international application, applications for patent or inventor's certificate on this invention of discovery which have been filed by me or my legal representatives or assigns in any country foreign to the United States of America are as follows:**

(a) none filed more than 12 months prior to said international application, unless named below:

(b) earliest filed less than 12 months prior to said international application (the priority of which is hereby claimed under 35 U.S.C. Section 365):

I hereby claim the benefit under Title 35, United States Code, §120, of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose information material to patentability as defined in Title 37, Code of Federal Regulations, §1.56, which occurred between the filing date of the prior application and the national or PCT international filing date of this application.

(Application Serial No.) (Filing Date) (Status)(patented, pending, abandoned)

(15)

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I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

The undersigned hereby authorizes the U.S. attorney or agent named herein to accept and follow instructions from _____ as to any action to be taken in the Patent and Trademark Office regarding this application without direct communication between the U.S. attorney or agent and the undersigned. In the event of a change in the persons from who instructions may be taken, the U.S. attorney or agent named herein will be so notified by the undersigned.

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